Supplementary Information: Particle Entrainment in Dead-End Pores by Diffusiophoresis

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I. SIMULATION DETAILS

A. Brinkman Approximation Simulation with MATLAB

We seek to solve the quasi-three-dimensional, incompressible Navier-Stokes equations as prescribed by the Brinkman approximation:

$$\rho\frac{\partial\overline{\mathbf{u}}}{\partial t} = -\boldsymbol{\nabla}_\perp p + \mu\nabla_\perp^2\overline{\mathbf{u}} - \frac{\mu}{k}\overline{\mathbf{u}}, \tag{1}$$

where $\overline{\mathbf{u}}$ is the depth-averaged fluid velocity, μ is the viscosity of the particle-laden electrolyte solution (approximately that of water), ρ is the fluid density, $k = \ell^2/12$ is the effective permeability of the medium which depends upon the channel depth, ℓ is the channel depth (here $\ell = 10$ μm), $\boldsymbol{\nabla}_\perp$ is the gradient in the direction perpendicular to the channel height (here $\boldsymbol{\nabla}_\perp = \hat{\boldsymbol{x}}\,\partial/\partial x + \hat{\boldsymbol{y}}\,\partial/\partial y$ as illustrated in Figure 1), and p is the pressure [1, 2]. We introduce the time-dependent term in Equation (1) because we adopt an artificial time-stepping approach to determine the steady-state flow. In order to solve Equation (1), we adopt an incompressible vorticity/streamfunction formulation [3]. We recall that the vorticity in the depth-wise direction (ω) is defined as:

$$\omega = \frac{\partial\overline{u}_y}{\partial x} - \frac{\partial\overline{u}_x}{\partial y}, \tag{2}$$

where $\overline{\mathbf{u}} = (\overline{u}_x, \overline{u}_y)$ is the depth-averaged velocity expressed in component form, and that the streamfunction (ψ) is expressed as:

$$\overline{u}_x = \frac{\partial\psi}{\partial y}, \tag{3a}$$

$$\overline{u}_y = -\frac{\partial\psi}{\partial x}. \tag{3b}$$

Thus, the recast form of Equation (1) is:

$$\frac{\partial\omega}{\partial t} = \nu\nabla_\perp^2\omega - \frac{\nu}{k}\omega, \tag{4}$$

where $\nu = \mu/\rho$ is the kinematic viscosity.

In MATLAB, we use Equation (4) to solve for the vorticity in time. Moreover, we use the fact that

$$\nabla_\perp^2\psi = -\omega \tag{5}$$

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to solve for the streamfunction in time by successive over-relaxation [3]. Given the expressions in Equation (3), we determine the fluid velocity from the streamfunction. We impose the following boundary conditions on the fluid velocity from which we derive the corresponding boundary conditions on the vorticity and streamfunction:

$$\overline{u}_x = 0 \quad \text{and} \quad \overline{u}_y = 0 \quad \text{at all walls with the exception of the lateral pore walls,} \tag{6a}$$

$$\overline{u}_x = 0 \quad \text{and} \quad \overline{u}_y = -\Gamma_w \frac{\partial \ln C}{\partial y} \quad \text{at the lateral pore walls,} \tag{6b}$$

$$\overline{u}_x = \frac{Q}{\ell w_c}\left(\frac{-3}{1-3A}\right)\left[-\frac{1}{3} + \frac{32}{\pi^4}\sum_{n,\,odd}^{\infty}\frac{\cosh\left(n\pi\frac{y}{\ell} - \frac{n\pi w_c}{2\ell}\right)}{n^4\cosh\left(\frac{n\pi w_c}{2\ell}\right)}\right] \quad \text{and} \tag{6c}$$

$$\overline{u}_y = 0 \quad \text{at the main channel inlet/outlet,}$$

where Γ_w is the diffusioosmotic mobility along the channel walls, w_c is the main channel width in the y-direction (here $w_c = 200$ μm), C is the solute concentration, Q is the injection flow rate at the inlet, and $A = \sum_{n,\,odd}^{\infty}\frac{64\ell}{\pi^5 n^5 w_c}\tanh\left(\frac{n\pi w_c}{2\ell}\right)$. We assume that the velocity at the inlet and outlet of the main channel corresponds to a depth-averaged, fully developed Poiseuille flow [4]. We also assume an initial condition of zero fluid velocity. Since the flow velocity is quasi-steady with respect to the evolving solute concentration, we use a nested time loop to update the solute concentration over several time steps (while holding the vorticity/streamfunction constant) according to the following advection-diffusion equation:

$$\frac{\partial C}{\partial t} + \overline{\mathbf{u}} \cdot \boldsymbol{\nabla}_\perp C = D_s \nabla_\perp^2 C, \tag{7}$$

where D_s is the effective diffusivity of the salt, and subject to the boundary conditions:

$$C = C_{\text{in}} \qquad \text{at the main channel inlet,} \tag{8a}$$

$$\hat{\boldsymbol{n}} \cdot \boldsymbol{\nabla}_\perp C = 0 \qquad \text{at the walls and at the outlet,} \tag{8b}$$

where $C_{\text{in}} = 0.1$ mM is the fixed, uniform solute concentration at the inlet and $\hat{\mathbf{n}}$ is the outward unit normal on each respective boundary. The initial solute concentration is set to $C_1 = 0.1$ mM in the main channel and $C_2 = 10$ mM within the dead-end pore. Subsequently, we update the vorticity/streamfunction and iterate [5]. We then determine the new particle position (whose initial position is specified and which is advected as a point particle) by integrating the local particle velocity $\mathbf{u} = \overline{\mathbf{u}} + \Gamma_d \boldsymbol{\nabla}_\perp \ln C$, where Γ_d is the diffusiophoretic mobility of the particle.

B. Three-Dimensional Simulation with OpenFOAM

In OpenFOAM, we begin by solving the three-dimensional, steady-state, incompressible Navier-Stokes equation given by

$$\rho\left(\mathbf{u}_f \cdot \boldsymbol{\nabla}\mathbf{u}_f\right) = -\boldsymbol{\nabla}p + \mu\nabla^2\mathbf{u}_f, \tag{9}$$

where $\mathbf{u}_f$ is the fluid velocity and both the fluid density, ρ, and viscosity, μ, are assumed to be constant. Whereas with the Brinkman approximation simulation in MATLAB we used an artificial time approach to find the steady-state, here we directly use an iterative steady-state solver based on the SIMPLE algorithm [6]. We impose the following boundary conditions:

$$\mathbf{u}_f = \bar{U}\hat{\boldsymbol{e}}_x \quad \text{and} \quad \hat{\boldsymbol{n}} \cdot \boldsymbol{\nabla}p = 0 \qquad \text{at the main channel inlet,} \tag{10a}$$

$$\mathbf{u}_f = -\Gamma_w \boldsymbol{\nabla}\ln C \quad \text{and} \quad \hat{\boldsymbol{n}} \cdot \boldsymbol{\nabla}p = 0 \qquad \text{at the walls,} \tag{10b}$$

$$\hat{\boldsymbol{n}} \cdot \boldsymbol{\nabla}\mathbf{u}_f = 0 \quad \text{and} \quad p = 0 \qquad \text{at the main channel outlet,} \tag{10c}$$

where $\bar{U}$ is the average velocity across the main channel cross-section (determined by dividing the injection flow rate by the cross-sectional area of the main channel), $\hat{\boldsymbol{e}}_x$ is the unit normal in the x-direction, $\hat{\boldsymbol{n}}$ is the outward unit normal on each respective boundary, C is the solute concentration, and Γ_w is the diffusioosmotic mobility along the channel walls. Upon solving for the (quasi) steady-state fluid velocity and pressure profiles, we update the solute concentration by integrating an advection–diffusion equation given by

$$\frac{\partial C}{\partial t} + \boldsymbol{\nabla} \cdot (\mathbf{u}_f C) = D_s \nabla^2 C, \tag{11}$$

where D_s is the effective diffusivity of the salt, and subject to the following boundary conditions:

$$C = C_{\text{in}} \qquad \text{at the main channel inlet,} \tag{12a}$$

$$\hat{\boldsymbol{n}} \cdot \boldsymbol{\nabla} C = 0 \qquad \text{at the walls and at the outlet,} \tag{12b}$$

where $C_{\text{in}} = 0.1$ mM is the fixed, uniform solute concentration at the inlet. As per Equation (12b), we assume that the solute does not penetrate the channel walls and that the flow is fully developed at the main channel outlet. The initial solute concentration is set to $C_1 = 0.1$ mM in the main channel and $C_2 = 10$ mM within the dead-end pore. Once the solute concentration is updated, we determine the new particle position (whose initial position is specified and which is advected as a point particle) by integrating the local particle velocity $\mathbf{u} = \mathbf{u}_f + \Gamma_d \boldsymbol{\nabla} \ln C$, where Γ_d is the diffusiophoretic mobility of the particle.

II. SUPPLEMENTARY FIGURES

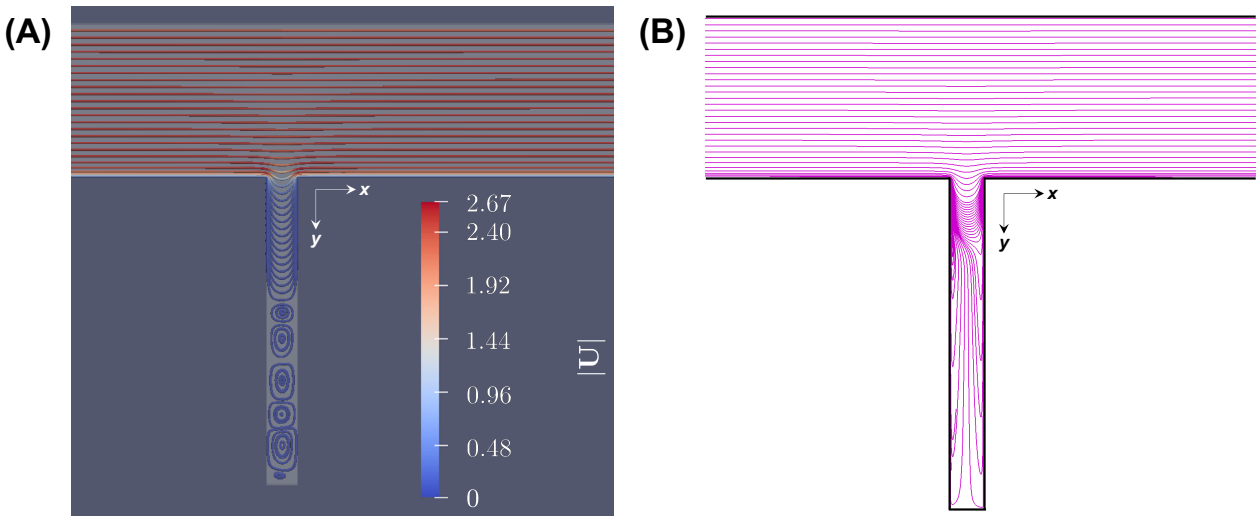

FIG. 1. Fluid streamlines simulated in three-dimensions with OpenFOAM. The dead-end pore is 40 μm in width and 400 μm in length, while the main channel is 200 μm wide; the entire device is 10 μm in depth. (A) In the absence of a solute concentration gradient, the flow lines along the center-plane (in the depth-wise direction) of the device reveal deeply penetrating streamlines. The magnitude of the fluid velocity scaled by the average velocity across the main channel cross-section, denoted by the non-dimensional variable ($|\mathbf{U}|$), is quantified by the color legend. (B) A recirculation current due to diffusioosmosis and pressure-driven flow within the pore arises in the presence of an electrolyte gradient. These streamlines along the center-plane of the device are recorded ten seconds after the commencement of flow through the main channel.

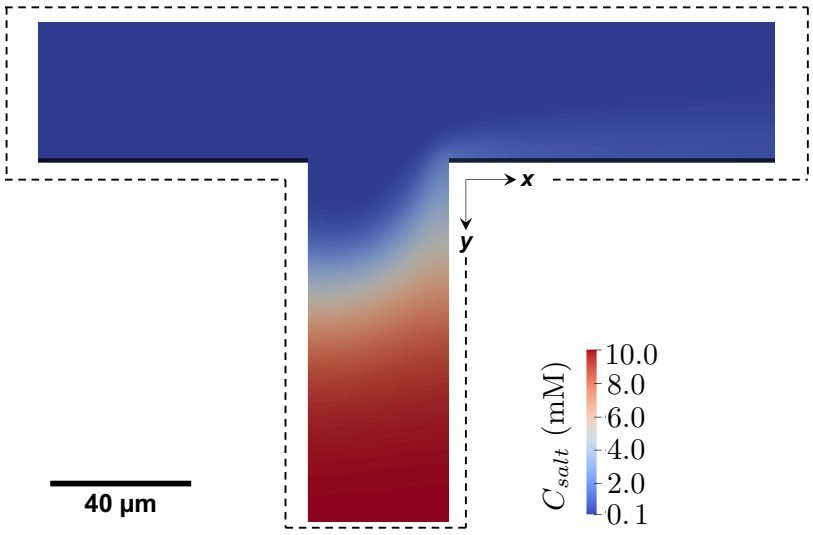

FIG. 2. Solute concentration profile in the dead-end pore geometry (zoomed-in view with full dimensions indicated in Figure 1 of the main text). Results from the OpenFOAM simulation of the electrolyte solution's concentration profile at $t = 0.25$ seconds after the commencement of the experiment and at a Reynolds number of $\mathrm{Re} = \mathcal{O}(0.01)$ indicate a lateral solute concentration gradient at the pore opening. The details of the simulation are described in Section I.B of the Supplementary Information section, where $D_s = 1.61 \times 10^{-9}$ m^2/s, $\Gamma_w = 1.90 \times 10^{-10}$ m^2/s, and $\Gamma_d = 6.64 \times 10^{-10}$ m^2/s. Since the Peclet number is of order $\mathrm{Pe} = \mathcal{O}(20)$ at the pore entry, advection dominates diffusion and sweeps fluid from the pore. The commencement of the experiment is the time at which the 0.1 mM electrolyte solution begins to flow through the main channel.

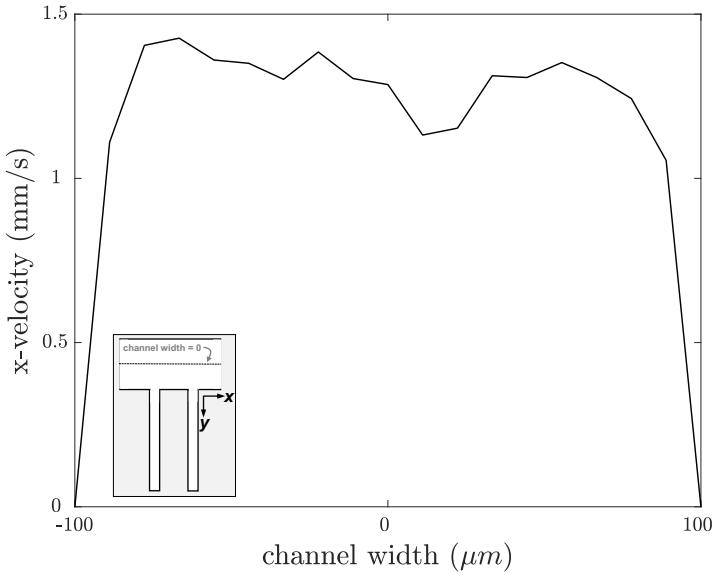

FIG. 3. Experimentally measured velocity profile of one-micron diameter particles flowing in the main channel with a depth of 10 μm at Re = 0.028 past a series of dead-end pores and subject to a solute concentration gradient. The velocity field is established from particle image velocimetry (PIV) analysis and evidences a rapid velocity decay near the channel walls. (inset) Schematic of the device, where the channel center (corresponding to a channel width of zero) is specified by the dotted line and the out-of-plane depth of the device is 10 μm.

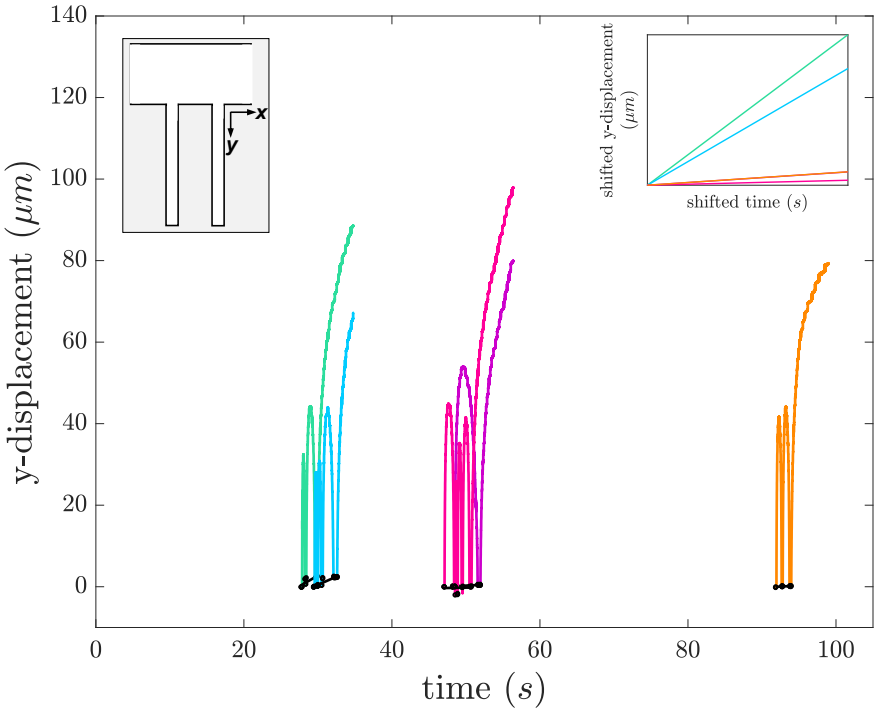

FIG. 4. Persistence in time of the pore-to-pore hopping pattern before particle capture. Plot of y-displacements of many one-micron diameter particles as a function of the experimental duration at Re = 0.042, where the displacement is measured relative to a particle's initially-tracked position and the y-direction is oriented along the pore's length. As evidenced by the orange particle trajectory, a particle tracked more than ninety seconds after the commencement of the experiment executes pore-to-pore hopping before eventual capture. (inset) Magnified view of the lines connecting the plateau regions (denoted by black circles in the main panel) of each particle trajectory, which are shifted to the origin and color-matched according to the color of the corresponding trajectory.

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[5] The time step is on the order of the minimum of the advective and solute diffusion timescales.
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